Journal of Physical Chemistry A 2008 vol.112 N15, pages 3291-3301

Correlation of transverse and rotational diffusion coefficient: A probe of chemical composition in hydrocarbon oils

Mutina A., Hürlimann M.
Kazan Federal University, 420008, Kremlevskaya 18, Kazan, Russia

Abstract

Measurements of relaxation time and diffusion coefficient by nuclear magnetic resonance are well-established techniques to study molecular motions in fluids. Diffusion measurements sense the translational diffusion coefficients of the molecules, whereas relaxation times measured at low magnetic fields probe predominantly the rotational diffusion of the molecules. Many complex fluids are composed of a mixture of molecules with a wide distribution of sizes and chemical properties. This results in correspondingly wide distributions of measured diffusion coefficients and relaxation times. To first order, these distributions are determined by the distribution of molecular sizes. Here we show that additional information can be obtained on the chemical composition by measuring two-dimensional diffusion-relaxation distribution functions, a quantity that depends also on the shape and chemical interactions of molecules. We illustrate this with experimental results of diffusion-relaxation distribution functions on a series of hydrocarbon mixtures. For oils without significant amounts of asphaltenes, the diffusion-relaxation distribution functions follow a power-law behavior with an exponent that depends on the relative abundance of saturates and aromatics. Oils with asphaltene deviate from this trend, as asphaltene molecules act as relaxation contrast agent for other molecules without affecting their diffusion coefficient significantly. In waxy oils below the wax appearance temperature a gel forms. This is reflected in the measured diffusion-relaxation distribution functions, where the restrictions due to the gel network reduce the diffusion coefficients without affecting the relaxation rates significantly.

http://dx.doi.org/10.1021/jp710254d